



RESEARCH MEMORANDUM

TRANSPORT OF RADIOACTIVITY BY LIQUID SODIUM
IN A STAINLESS STEEL CIRCULATION SYSTEM

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STAINLESS STEEL CIRCULATION SYSTEM

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SUMMARY

Radioactivity transport experiments were conducted in a stainless steel system in which liquid sodium was circulated over an activated stainless steel insert section. The system consisted of a sealed toroid loop partly filled with sodium. Circulation of the sodium was induced by an oscillatory motion of the toroid in an apparatus developed at the NACA Lewis laboratory. Periodic measurements of radioactivity at sections of the toroid loop established the growth of transported activity in the loop.

The experimental evidence obtained substantiated a qualitative explanation that activity transfer in out-of-pile isothermal sodium - stainless steel systems is limited by diffusion of activated atoms in stainless steel. The radioactivity transferred from the active insert to the toroid wall increased with increase in sodium oxide concentration in the sodium. Data were obtained from three toroids operated at a constant wall temperature of 900° F and a constant sodium velocity of 15 feet per second.

One of the toroid loops, previously operated 1400 hours at isothermal conditions, was operated with temperature differentials obtained by cooling a small section of the toroid to check for mass-transfer effects. No further change in the previously established equilibrium isothermal value of transported activity was found after 600 hours at temperature differential conditions, which provides additional evidence that the limiting process in an out-of-pile experiment is the diffusion of active atoms in the stainless steel.

INTRODUCTION

The problem of transport of radioactive constituents of stainless steel by liquid sodium arises in connection with the Submarine Intermediate Reactor (SIR) and accessibility at sea to compartments surrounding the reactor. In order to investigate this problem of surface contamination in circulating liquid sodium systems, a Radioactive Accessibility Committee was formed. NACA has participated as a member of this Committee in cooperation with the Naval Reactors Branch of the AEC, Knolls Atomic Power Laboratory (KAPL), and other industrial organizations and government agencies.

Radioactive constituents of stainless steel available for transport from a reactor to other parts of a circulating system arise from two sources: (1) continuous activation of atoms in the stainless steel fuel elements and (2) direct activation of stainless steel atoms dissolved in sodium in passing through the reactor. Both of these sources cannot be reproduced in an out-of-pile experiment. The first source cannot be completely simulated in an out-of-pile experiment because the fuel element surfaces are gradually depleted of active atoms whereas in an in-pile experiment, atoms near the surface are constantly being activated. However, one aspect of the first effect can be studied by operation of an out-of-pile loop with an activated section. Accordingly, the transport of radioactivity from an activated stainless steel insert section to the inside surface of the circulating loop was measured as a function of time at approximately SIR operating conditions. The effects of oxide content, velocity, and temperature of sodium, and surface temperature of stainless steel on the magnitude of activity transport were determined.

The activity transport experiments performed by NACA have used an oscillating toroid apparatus described in reference 1 in which desired system temperatures and sodium velocities may be simulated. This experimental system was used for the activity transport problem because the toroids require no pump, are relatively easy to fabricate, and can be filled and sealed from the atmosphere for the entire period of the experiment.

Preliminary data and a description of the apparatus and counting techniques were presented in reference 2. The results of the completed experimental program are presented herein. Isothermal results and results for a toroid with temperature differentials maintained between a heated and a cooled toroid section have been obtained.

EXPERIMENTAL ARRANGEMENT

Circulation apparatus. - A photograph of the circulation apparatus for isothermal operation of toroid loops is shown in figure 1. The

toroid loop is located inside an electrically heated furnace and mounted on an oscillating plate to induce circulation of molten sodium. The furnace permits easy insertion and removal of a toroid during periodic shutdown for measurement of transferred radioactivity. The circulation apparatus is more fully described in references 1 and 2.

For operation with a temperature differential, two of the twelve heating elements were removed from one section of the furnace. Forced circulation of air over this section provided a cooled region on the toroid surface.

Toroids. - Each toroid was rolled from 1/2-inch-inside-diameter AISI type 347 stainless steel tubing with 1/16-inch walls. The welded assembly of the 15 $\frac{1}{2}$ -inch-diameter toroid and an AISI type 347 stainless steel collar housing a radioactive insert is illustrated in figure 2. The toroid also included two small vent tubes for flushing and loading with sodium which were sealed off after the loading was completed. Toroids were provided by NACA; cleaning and filling of the toroids were performed by KAPL. Each toroid was degreased with hot trisodium phosphate cleaner, leak-tested with a mass spectrometer, and then further cleaned by flushing with 625° F sodium at 0.6 gallon per minute for 24 hours. After the toroid had been cleaned, it was loaded to about 40 percent of capacity with sodium of known oxygen content. Table I lists pertinent data concerning the three toroids tested.

For isothermal operation, each toroid was instrumented with six equally spaced chromel-alumel thermocouples: one for furnace temperature control located near the active insert and five for continuous temperature recording. For temperature differential operation of toroid 1, ten chromel-alumel thermocouples were used: four in the cooled section, and six in the hot section of the toroid. The thermocouple located near the active insert in the hot section was again used for furnace temperature control. One control thermocouple was sufficient to keep the temperature within $\pm 5^\circ$ F of the desired toroid wall temperature for both the isothermal and temperature differential measurements. The thermocouples were spot-welded to the periphery of the toroid. The thermocouples in the cold section were protected from the direct air blast by a commercial cement covering.

Radioactive inserts. - The inserts for toroids 1 and 2 were made of 3.3-mil AISI type 316 stainless steel foil. After irradiation, the insert was spot-welded to the inner surface of the collar shown in figure 2. The foil was irradiated at Brookhaven National Laboratory for 100 days at an average thermal flux of 2.38×10^{12} neutrons per square centimeter per second. Radiochemical and chemical analysis of the irradiated 316 stainless steel foil as furnished by KAPL is given in table I. Table I indicates that Cr⁵¹ and Fe⁵⁹ contribute an appreciable part

of the activity of the foils. At the time of the experiments with toroids 1 and 2, however, the activities of the relatively short-lived atoms of Cr^{51} and Fe^{59} had effectively decayed out, leaving only the Co^{60} activity.

The active insert in toroid 3 was made from the same stock of AISI type 347 stainless steel used in making toroid 3. The tubing was turned on a lathe to a thickness of 6 mils and a length of 5/8 inch. This insert was irradiated at Brookhaven National Laboratory for 61 days at an average thermal flux of 3.14×10^{12} neutrons per square centimeter per second. A radiochemical analysis furnished by KAPL is given in table I; a nominal chemical analysis is included in the table. AISI type 347 stainless steel contains some tantalum which is a hard gamma emitter. The two relatively long-lived active constituents of AISI type 347 stainless steel are therefore Co^{60} and Ta^{182} .

Shielded counting assembly. - The assembly used for measuring transported radioactivity is shown in figure 3. The lead-brick shielding used in the construction reduced the response of the detector to radiation from the primary radioactive insert to a point where this activity registered considerably less than the normal background. The scintillation counter and counter shield shown in figure 3 can be set in any of three positions over the toroid. These measuring positions are on the side opposite the radioactive insert. One position is directly opposite the insert and includes about 55° of toroid arc; the other positions are to both sides and also include about 55° of toroid arc. This can be clearly seen in figure 3 with the shielded scintillation counter placed in one of the counting positions.

A calibration of the counter, described in the appendix of reference 2, showed that over 90 percent of the radioactivity detected came from a section of the toroid about 9 centimeters long and directly beneath the counter. A section of toroid 9 centimeters long is about half the toroid arc covered by the shielded counter. The shielded counter reduced detectable background to an average of 0.25 count per second.

PROCEDURE

The activity transported by the sodium from the active insert to the walls of the toroid was measured as a function of operating time for each toroid. After the toroid was operated in the circulation apparatus for a specified length of time, it was removed from the furnace and the sodium allowed to drain and freeze in the section containing the active insert. Background and counter consistency measurements were made before the toroid was brought into the counting area; the consistency of the scintillation counter was checked with a Co^{60} source. The experimental data consisted of three counts taken at each of the three measuring positions of the toroid.

3470 The calibration described in the appendix of reference 2 was used to calculate the fraction of the total insert activity transported to a unit area of the toroid wall from these experimental data. Inasmuch as the activity available for transport to the toroid inner surface depends on the activity per unit volume of active insert, the data for various inserts may be put on a common basis by using activity per unit volume of active insert. Since the insert densities are very closely the same, activity per unit mass of insert serves as a more convenient measure. Therefore, the data are reported as specific transported activity f , defined as the fraction of the activity in a milligram of insert material transported and deposited on a unit area of the toroid wall.

RESULTS

Isothermal experiments. - The specific transported activity f , averaged for the three measuring positions, is plotted in figure 4 as a function of the total time at operating conditions. Data are presented for toroids 1, 2, and 3 operated at a constant temperature of 900° F and a sodium velocity of 15 feet per second. The three curves have the same general shape and indicate a gradual increase and final leveling off in activity transported with increasing operating time.

The variation observed in figure 4 may be explained by the assumption that the activity transport process is diffusion limited; that is, that diffusion of active atoms from within the active insert and into the toroid walls is a process which is relatively slow compared to the period of toroid operation. At the start of toroid operation, sodium dissolves both active and inactive atoms of the various constituents of stainless steel in proportion to their solubilities. During the period of toroid operation these dissolved active and inactive atoms undergo random exchanges with atoms on both the active insert and the toroid wall. This process results in a fast rise in activity deposited on the toroid wall during the early period of operation. As toroid operating time increases, the surface of the active insert becomes gradually depleted of active surface atoms if diffusion of active atoms from inner layers of the insert to the outer surface is a relatively slow process. Thus, after relatively long periods of toroid operation, as the surface of the active insert is effectively depleted, the specific transported activity shown in figure 4 gradually levels off. The activity transport process would therefore appear to be limited by diffusion in the active insert material. Similar conclusions were reached in reference 3.

Additional experimental evidence for this qualitative explanation was obtained with toroid 3. In addition to the usual measurements, periodic measurements of toroid 3 with the sodium frozen in the measured section opposite the active insert were made to measure the amount

of activity present in the volume of sodium. The variation with time of the total activity in the sodium and on the toroid walls and the activity on the toroid walls only is shown in figure 5. The activity in the sodium only, obtained by difference, is also shown in figure 5 as a function of operating time. This curve indicates that the concentration of active atoms in the sodium reaches a maximum shortly after startup and gradually decreases after relatively long periods of isothermal operation. These data are in agreement with the qualitative explanation offered previously and indicate further that the activity transport process in the toroid loops is diffusion limited.

The relative magnitude of the curves in figure 4 are presumed to be due principally to the effect of oxygen (sodium oxide) content of sodium. The adverse effect of oxygen on corrosion by liquid metals is well known. A comparison of the activities transported at the end of 650 hours of isothermal operation at 900° F is listed for the three toroids with different oxygen content in table I. Qualitatively, the data indicate that an increase in the oxygen content of sodium is accompanied by an increase in the radioactivity transported.

The effect of an increase in toroid operating temperature on the magnitude of isothermal activity transport was measured with toroid 1. This toroid was operated at 1050° F and flow velocity of 15 feet per second. These results are included in figure 6 and compared with the data at 900° F and flow velocity of 15 feet per second. Before the test at the higher temperature was begun, the activity on the toroid walls was again counted. This activity is indicated as the check point on the figure at 1092 hours. Upon putting the toroid into operation at 1050° F, it was found that the activity on the toroid walls abruptly decreased about 10 percent at all three measuring stations and gradually increased during 300 hours of subsequent operation at 1050° F. An explanation is that at the higher operating temperature, the increased solubility of constituent atoms of stainless steel in sodium resulted in removal of some of the active atoms deposited on all surfaces of the toroid wall. After a period of isothermal operation at the higher temperature, during which random exchange of dissolved and surface atoms occurred, a new equilibrium was established. The higher equilibrium value of specific transported activity indicated on figure 6 would reflect the effect of greater diffusivity in the active insert at higher temperature.

Temperature differential experiments. - Previous work (ref. 4) had indicated that dynamic corrosion effects such as mass transfer were negligible in a type 347 stainless steel sodium system operated at 1500° F with a cooled section at 1460° F. These findings were checked by operation of toroid 1 with heated and cooled sections at various temperatures. The greater activity transported isothermally in toroid 1 made it desirable for measuring the effects of temperature differentials on the magnitude of activity transport. Toroid 1 was first operated for

400 hours with a maximum temperature differential of 25° F with the outside toroid wall temperature in the hot section at 900° F and a sodium flow velocity of 15 feet per second. A schematic diagram of toroid 1 with outside wall temperatures and direction of sodium flow is shown in figure 7.

3470 Following the 400 hours of operation, a new temperature differential condition was investigated for 75 hours: hot section maintained at 750° F with a maximum temperature differential in the cold section of 60° F and a sodium flow velocity of 15 feet per second. This in turn was followed by 100 hours of operation with the hot section at 850° F, a maximum temperature differential in the cold section of 70° F, and a flow velocity of 10 feet per second. The results of these tests are summarized in figure 8 in which the data for the three counted sections of the toroid are separately indicated. No marked change in specific transported activity f above the isothermal value was observed after a total of nearly 600 hours of operation at three temperature differential conditions. These results must be interpreted in the light of findings of the isothermal experiments. The isothermal data of figure 6 indicate that after 1400 hours of operation, the activity transport process in toroid 1 was mainly dependent on the diffusion rate of active atoms in the activated insert. The surface of the insert source had by this time been largely depleted of active atoms. In putting toroid 1 into operation with a temperature differential, the hot temperature was reduced from its isothermal operating value of 1050° F with a corresponding reduction in the diffusion rate. This would explain the small increase in transported activity observed in figure 8. It is possible that mass-transfer effects would be more pronounced at earlier stages of the experiment or with an in-pile experiment where surfaces were being continuously reactivated.

Although these data indicate that mass-transfer effects were small for these toroid operating conditions, slightly higher values of f were generally observed in the counting sections first contacted by the sodium after passing over the radioactive insert source.

SUMMARY OF RESULTS

Radioactivity transport experiments were conducted in a stainless steel system in which liquid sodium was circulated over an activated stainless steel insert section. The system consisted of a sealed toroid loop partly filled with sodium. Circulation of molten sodium was induced by an oscillatory motion of the toroid. The following results were obtained:

1. Experimental evidence substantiated a qualitative explanation that activity transfer in out-of-pile isothermal sodium - stainless steel systems is limited by diffusion of activated atoms in stainless steel.

2. Radioactivity transferred from the active insert to the toroid walls increased with increase in sodium oxide concentration in sodium as indicated by experiments with three toroids which were operated at a constant wall temperature of 900° F and a constant sodium velocity of 15 feet per second.

3. A toroid loop, with an operating history of 1400 hours at isothermal conditions, was operated with temperature differentials obtained by cooling a small section of the toroid. No further change in the isothermal value of transported activity was found after an additional 600 hours at temperature differential conditions. This is further evidence that in an out-of-pile experiment diffusion of atoms through the active section is the controlling factor in the transport of radioactivity.

Lewis Flight Propulsion Laboratory
National Advisory Committee for Aeronautics
Cleveland, Ohio, November 15, 1954

REFERENCES

1. Desmon, Leland G., and Mosher, Don R.: Preliminary Study of Circulation in an Apparatus Suitable for Determining Corrosive Effects of Hot Flowing Liquids. NACA RM E51D12, 1951.
2. Spooner, Robert B., and Fieno, Daniel: Preliminary Study of Transport of Radioactivity from Stainless Steel in Liquid-Sodium Circulation Systems. NACA RM E53K18, 1953.
3. Kendall, W. W.: Review of Stainless Steel Transport in SIR Radioactive Accessibility. Memo. CTU-7, Prog. Rep., Knolls Atomic Power Lab., Sept.-Oct., 1953.
4. Mosher, Don R., and Desmon, Leland G.: Dynamic Corrosion of AISI 347 Stainless Steel by Sodium at 1500° F. NACA RM E52G10, 1952.

TABLE I. - TOROID SPECIFICATIONS AND ACTIVE INSERT DATA

	Toroid		
	1	2	3
Toroid material	347 SS	347 SS	347 SS
Active insert material	316 SS	316 SS	347 SS
Sodium oxygen content ^a , percent by weight	(b)	0.003	0.005
Specific transported activity, f (isothermal 900° F, flow velocity 15 fps, 650 hr operation)	16.1×10^{-4}	0.8×10^{-4}	4.6×10^{-4}
Activity transported relative to toroid 2 for isothermal conditions	20.1	1.0	5.8
Active insert Radiochemical analysis ^c , d/min/g:			
Co ⁶⁰	2.82×10^9 (12/3/52)	2.82×10^9 (12/3/52)	1.60×10^9 (11/1/53)
Cr ⁵¹	$.91 \times 10^9$ (12/3/52)	$.91 \times 10^9$ (12/3/52)	8.00×10^8 (11/1/53)
Fe ⁵⁹	$.59 \times 10^9$ (12/3/52)	$.59 \times 10^9$ (12/3/52)	2.6×10^7 (11/1/53)
Ta ¹⁸²	-----	-----	5.2×10^9 (4/14/54)
Active insert Chemical analysis, percent by weight:			
Cr	17.26	17.26	d _{17.0}
Ni	12.55	12.55	d _{8.0}
Co	.106	.106	d _{.05}
Mn	-----	-----	d _{2.0}
Ta	-----	-----	d _{.01}
Fe	68.0 (by difference)	68.0 (by difference)	d _{70.0}
Date of isothermal operation at 900° F, flow velocity, 15 fps	3/24/53-6/25/53	6/23/53-9/14/53	10/19/53-2/8/54
Total isothermal operating time (hr)	1092	744	678

^aData provided by KAPL.^bLeak in filling toroid 1 estimated by KAPL to increase oxygen content to 0.009 percent from 0.0045 percent originally specified.^cData concerning activity on dates indicated provided by KAPL.^dNominal chemical analysis.



Figure 1. - Toroid circulation rig with toroid in furnace.

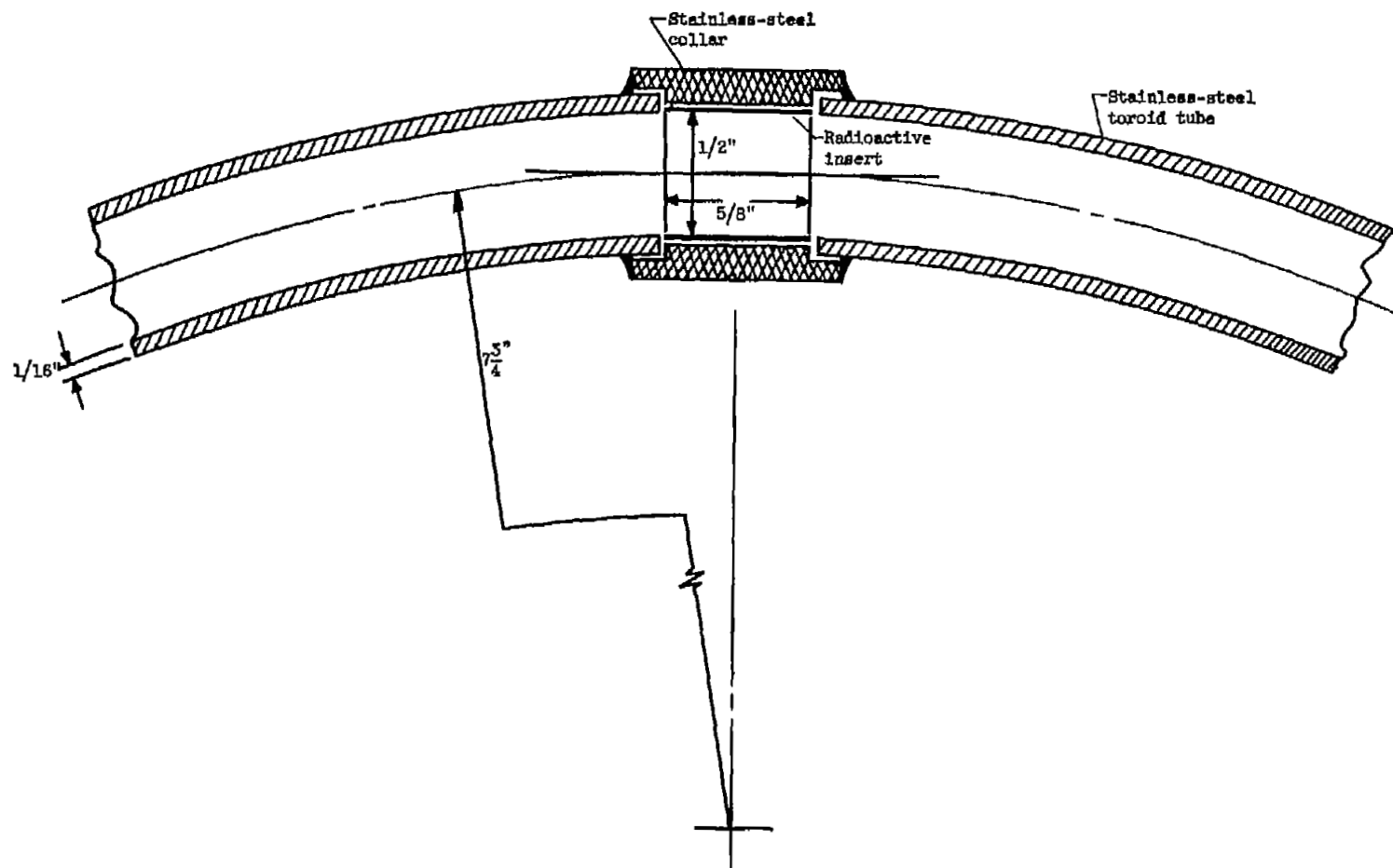


Figure 2. - Toroid assembly.

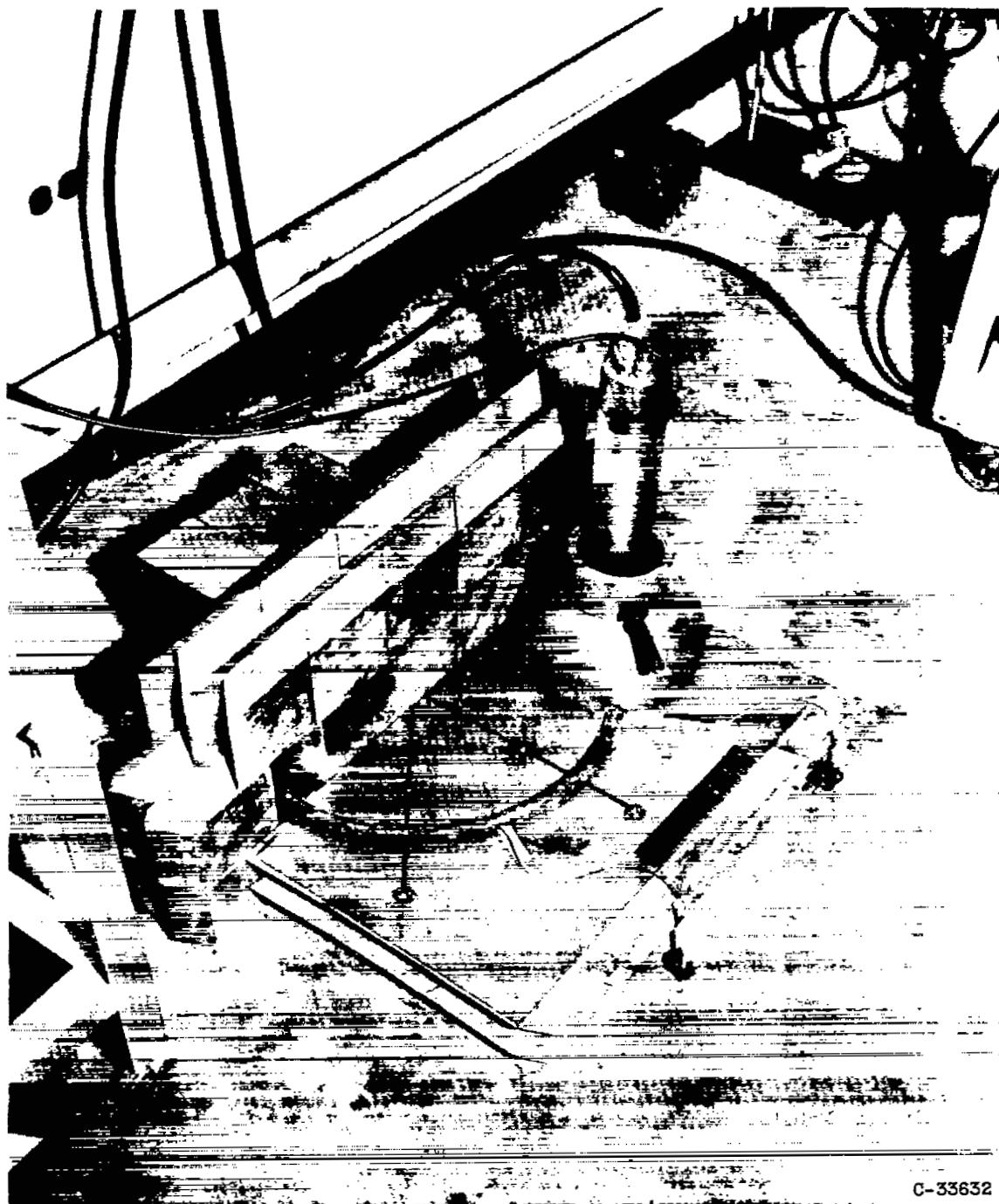


Figure 3. - Assembly for measuring transferred radioactivity.

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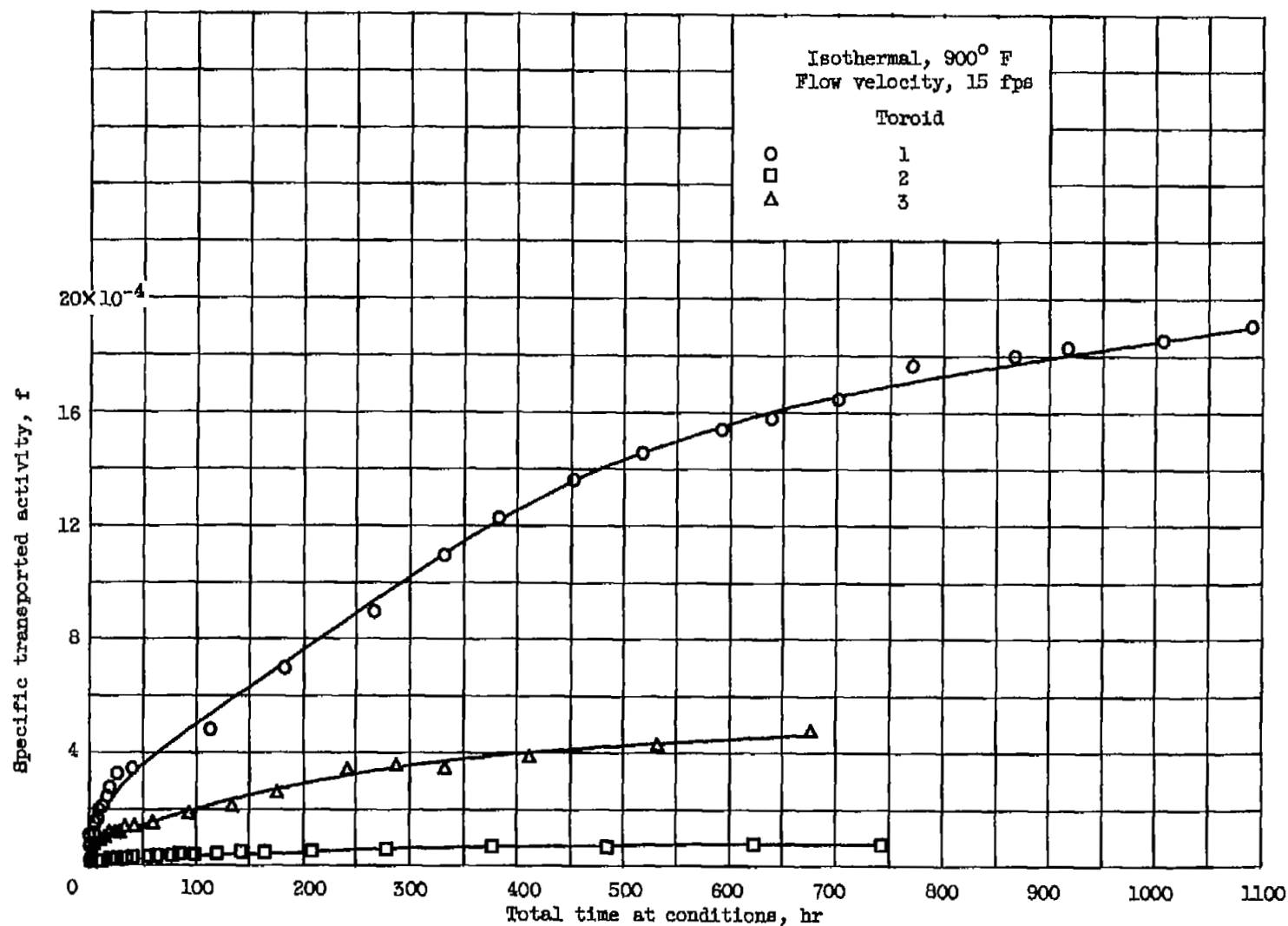


Figure 4. - Comparative isothermal activity transport data for three toroids.

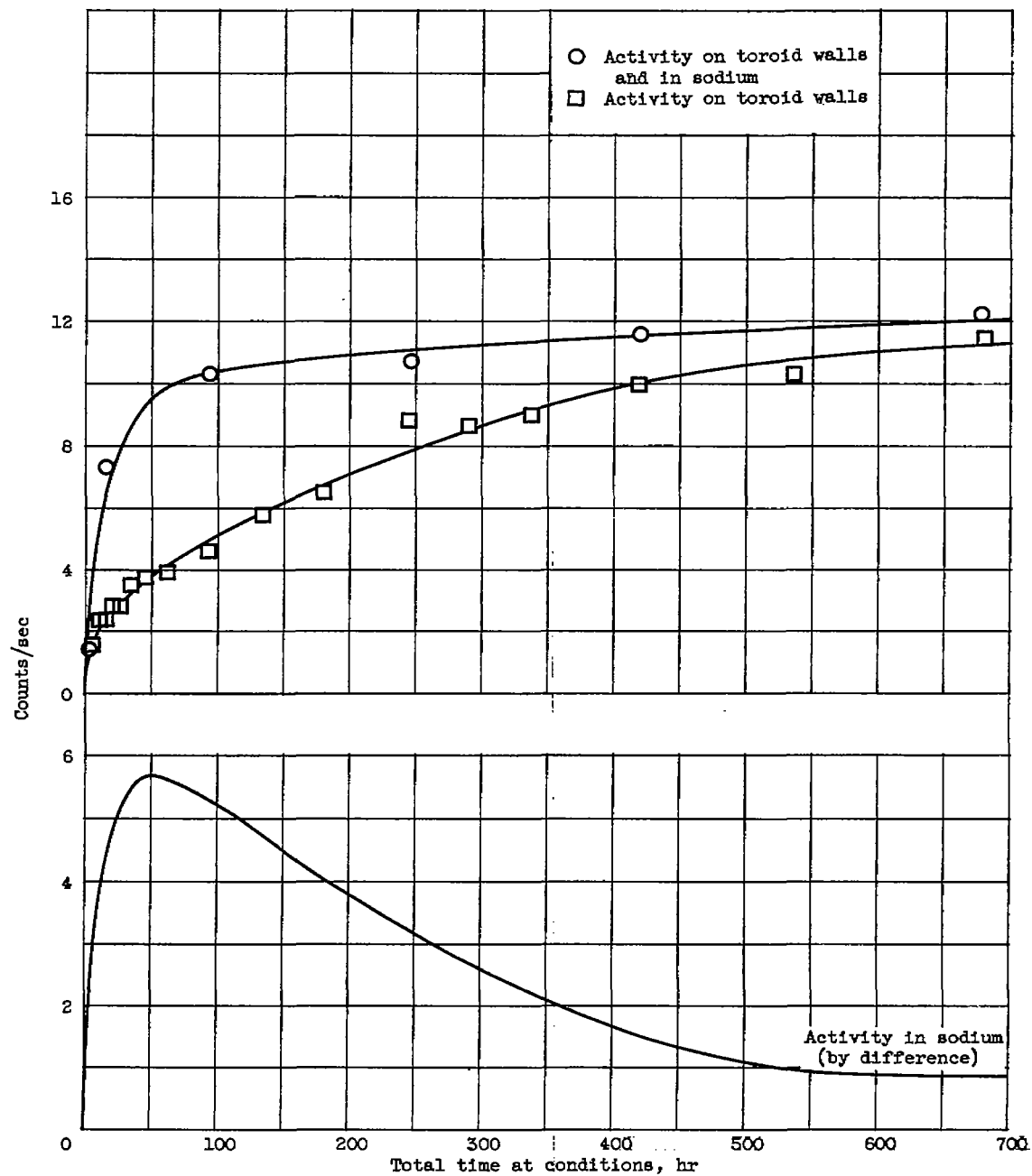


Figure 5. - Activity in sodium and on walls of toroid 3.

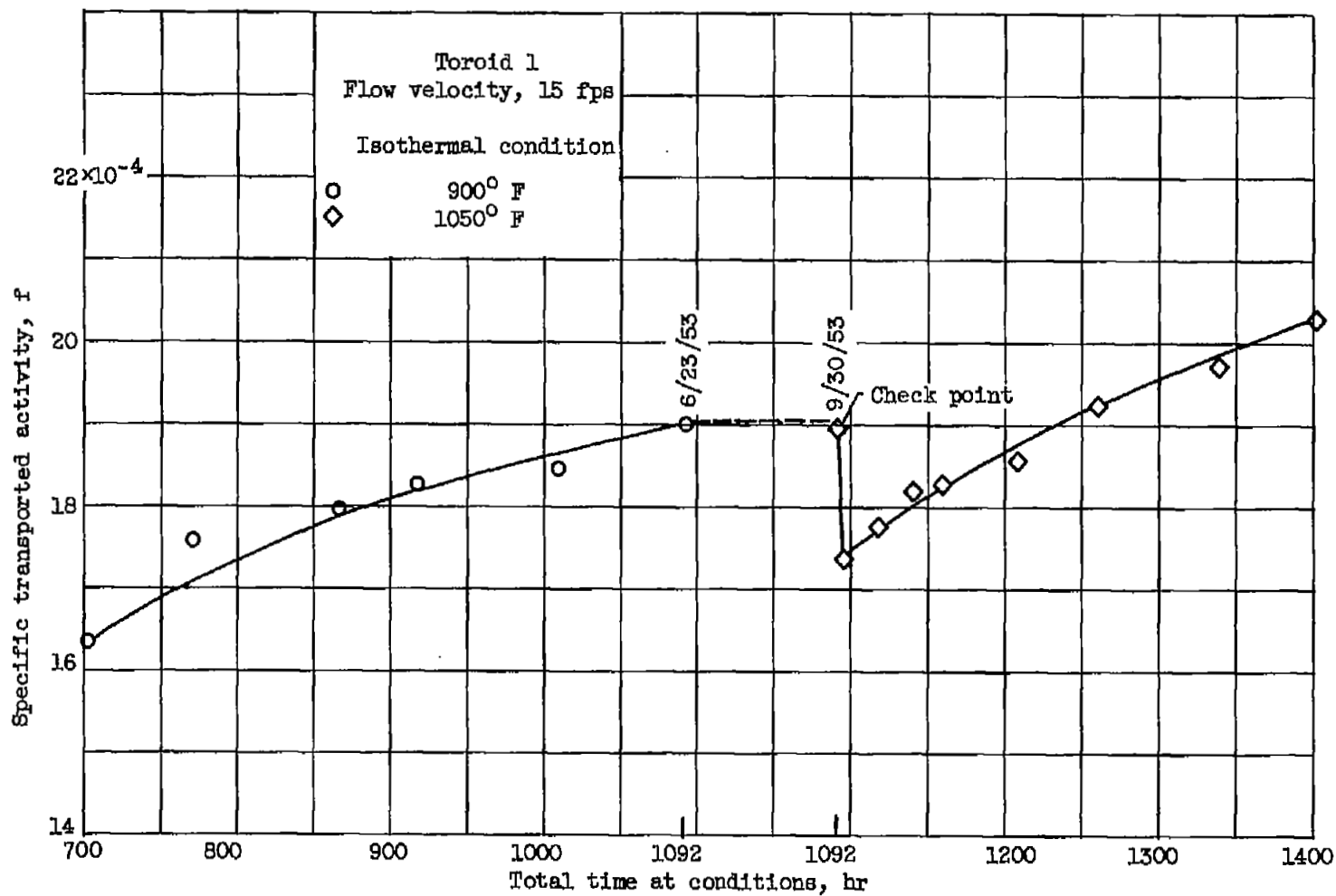


Figure 6. - Effect of change in isothermal operating temperature for toroid 1.

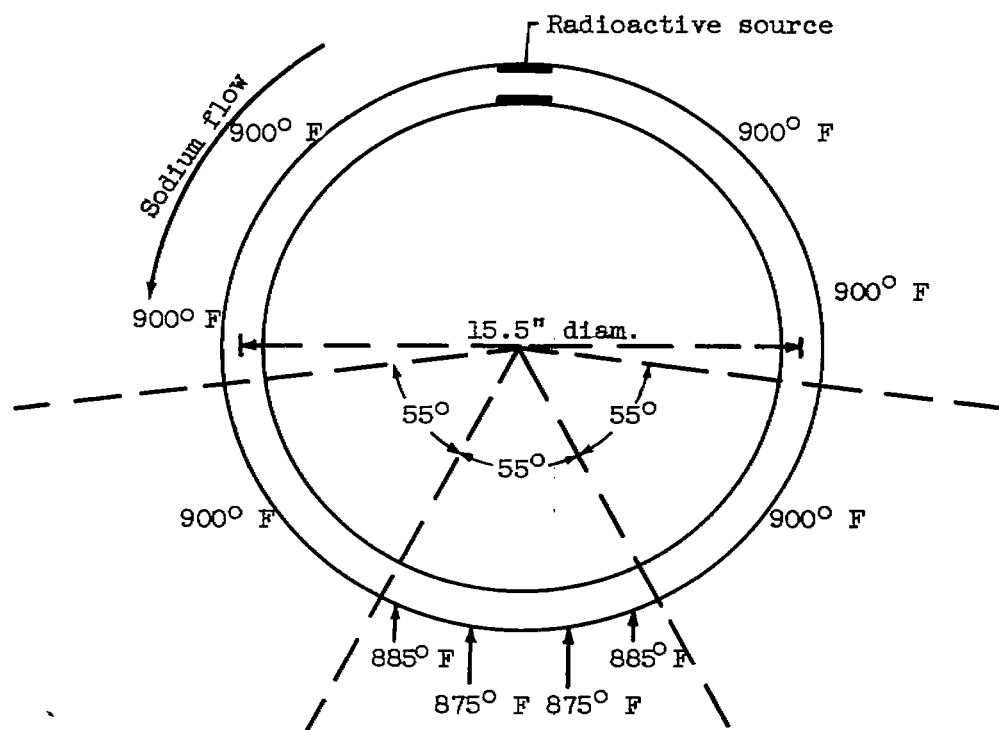


Figure 7. - Schematic diagram of toroid 1 showing temperature distribution and direction of sodium flow for a temperature differential condition.

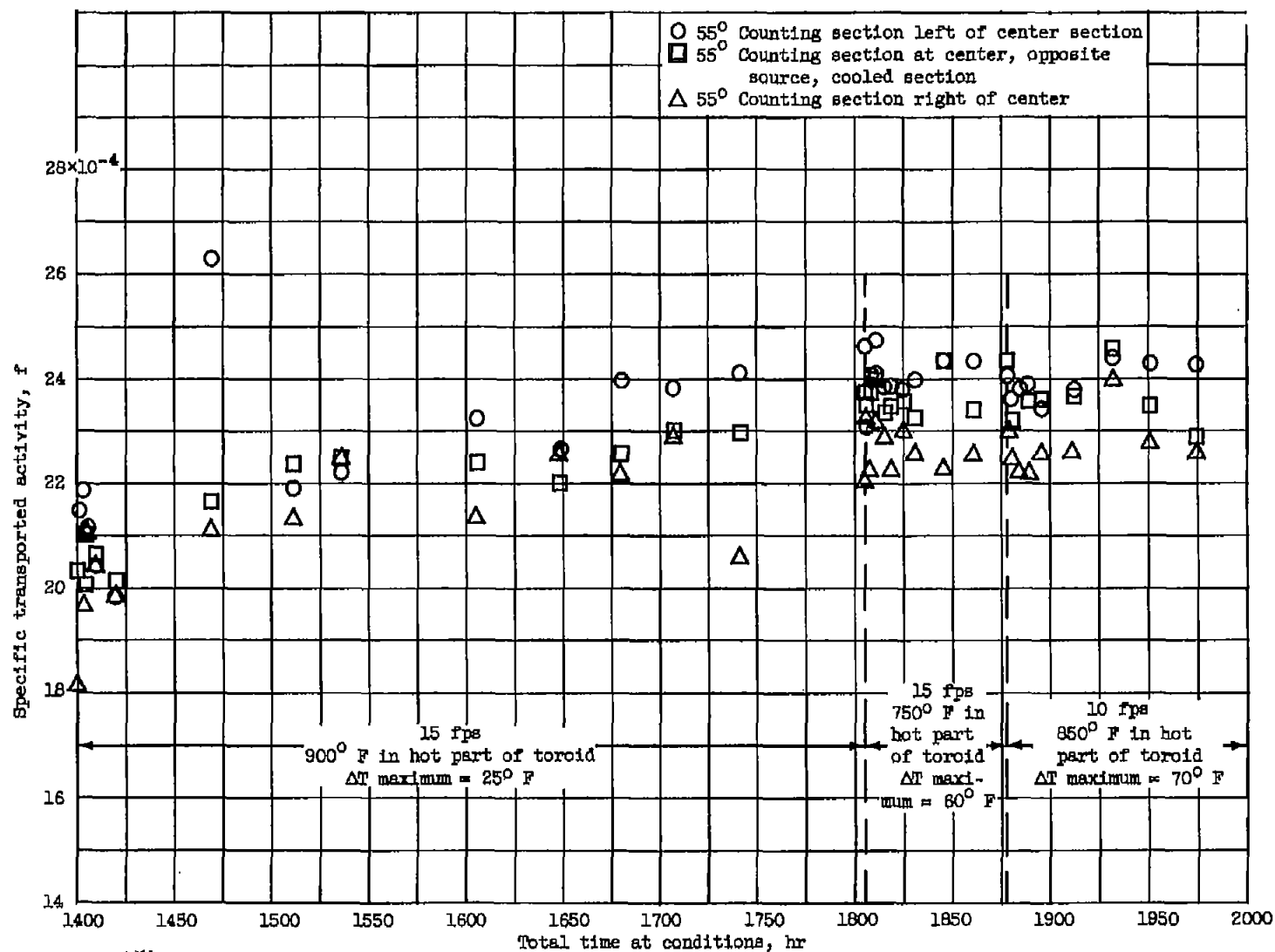


Figure 8. - Data for three counted sections of toroid 1 for three ΔT conditions.

